Express Mailino 740 168 US U S DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE ATTORNEY'S DOCKET NUMBER TRANSMITTAL LETTER TO THE UNITED STATES 5196-000003 DESIGNATED/ELECTED OFFICE (DO/EO/US) U.S APPLICATION NO. (If known, see 37 CFR 1.5 Ø CONCERNING A FILING UNDER 35 U.S.C. 371 890548 RNATIONAL APPLICATION NO. INTERNATIONAL FILING DATE PRIORITY DATE CLAIMED **₹₹**\$G00/00013 31 January 2000 (31.01.00) 02 February 1999 (2.02.99) TITLE OF INVENTION JEWELLERY ALLOY COMPOSITIONS APPLICANT(S) FOR DO/EO/US LOH, PENG CHUM Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information: 1. X This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371. This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below. The US has been elected by the expiration of 19 months from the priority date (Article 31). A copy of the International Application as filed (35 U.S.C. 371(c)(2)) is attached hereto (required only if not communicated by the International Bureau). Ъ. has been communicated by the International Bureau. is not required, as the application was filed in the United States Receiving Office (RO/US). An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). is attached hereto. International Application filed in English has been previously submitted under 35 U.S.C. 154(d)(4). Amendments to the claims of the International Aplication under PCT Article 19 (35 U.S.C. 371(c)(3)) are attached hereto (required only if not communicated by the International Bureau). have been communicated by the International Bureau. have not been made; however, the time limit for making such amendments has NOT expired. d. have not been made and will not be made. An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)). An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). An English lanugage translation of the annexes of the International Preliminary Examination Report under PCT 10. Article 36 (35 U.S.C. 371(c)(5)). Items 11 to 20 below concern document(s) or information included: An Information Disclosure Statement under 37 CFR 1.97 and 1.98. An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. A FIRST preliminary amendment. A SECOND or SUBSEQUENT preliminary amendment. 15. A substitute specification. A change of power of attorney and/or address letter. A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825. A second copy of the published international application under 35 U.S.C. 154(d)(4). A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4). Other items or information: Application Data Sheet, copy of 371 National Application of PCT/SG00/00013 (11 pages), copy of International Search Report, Form PTO-1449 with documents listed thereon (3 JP) and return postcard.

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| Harness, Dickey P. O. Box 828 | & Pierce, P.L | .C. | | Linda N | M. Deschere | |
| P. O. Box 828 Bloomfield Hills | Michigan 49 | 2303 | | NAME | | |
| United States of | | ,505 | | 34,811 | | |
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| | | | | Dated: | 7-31-01 | |

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Application No.:

Not yet assigned – 371 of PCT/SG00/00013

Filing Date:

Not yet assigned

Applicant:

LOH, Peng Chum

Title:

JEWELLERY ALLOY COMPOSITIONS

Attorney Docket:

5196-000003

Box Patent Applications Hon. Commissioner of Patents and Trademarks Washington, D.C. 20231

FIRST PRELIMINARY AMENDMENT

Sir:

Applicants herewith submit this First Preliminary Amendment to the application filed herewith, for consideration prior to the calculation of the filing fee, as follows:

IN THE CLAIMS

Please amend the claims in accordance with the following rewritten claims in clean form. Applicant includes herewith an Attachment for Claim Amendments showing a marked up version of each amended claim.

- 1. (AMENDED) A jewellery alloy comprising 76 83.5 wt% gold and 16.5 21.5 wt% aluminum, and having a substantially purple hue.
- 4. (AMENDED) A jewellery alloy according to claim 1, consisting of more than 78.5 wt% and up to and including 83.5 wt% gold and a balance of aluminum.

- (AMENDED) A jewellery allow according to claim 1, further comprising an additional element selected from the group consisting of palladium and nickel.
- 8. (AMENDED) A jewellery alloy according to claim 5, wherein the additional element is palladium and is present in an amount of between 0.5 wt% and 4.0 wt%.
- 9. (AMENDED) A jewellery alloy according to claim 5, wherein the additional element is nickel and is present in an amount of between 1.0 wt% and 2.0 wt%.
- 10. (AMENDED) An article comprising a metal component, wherein the metal component comprises a jewellery alloy according Claim 1.
- 13. (AMENDED) An alloy comprising 16.5 21.5 wt% aluminum, 0 4.0 wt% palladium, 0 2 wt% nickel and the balance gold.
- 14. (AMENDED) The alloy of claim 13 containing 18.5 19.5 wt% aluminum, 0.5 4.0 wt% palladium and the balance gold, except for impurities and incidental elements.
- 15. (AMENDED) The alloy of claim 13 containing 18.5 19.5 wt% aluminum, 1.0 2.0 wt% nickel and the balance gold, except for impurities and incidental elements.

REMARKS

Claims 1, 4, 5, 8 through 10, and 13 through 15 have been amended. Claims 1 through 15 remain pending in the application. The purpose of this Preliminary Amendment is to clarify the translation and remove multiple dependent claims from the application to reduce filing costs. It is submitted that this Amendment has antecedent basis in the application as originally filed, including the specification, claims and drawings, and that this Amendment does not add any new subject matter to the application. Consideration of the application as amended is requested. It is submitted that this Amendment places the application in suitable condition for allowance; notice of which is requested.

If for some reason Applicant has not paid a sufficient amount to prevent abandonment of this application, the Commissioner is hereby authorized to charge any fee due or credit any overpayment to deposit account no. 08-0750. A duplicate copy of this document is enclosed for this purpose.

If the Examiner feels that prosecution of the present application can be expedited by way of an Examiner's amendment, the Examiner is invited to contact the Applicant's attorney at the telephone number listed below.

Respectfully submitted,

Dated: 3/ July 200/

Linda M. Deschere Attorney for Applicants Registration No. 34,811

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Harness, Dickey & Pierce P. O. Box 828 Bloomfield Hills, MI 48303

ATTACHMENT FOR SPECIFICATION AMENDMENTS

The following is a marked up version of each replacement paragraph and/or section of the specification in which underlines indicates insertions and brackets indicate deletions.

- (AMENDED) A jewellery alloy [as hereinbefore defined,] comprising
 83.5 wt% gold and 16.5 21.5 wt% aluminum, and having a substantially purple
 hue.
- 4. (AMENDED) A jewellery alloy according to <u>claim 1</u> [any one of the preceding claims], consisting of more than 78.5 wt% and up to an including 83.5 wt% gold and a balance of aluminum.
- 5. (AMENDED) A jewellery allow according to [any one of] claim[s] 1[to 3], further comprising an additional element selected from the group consisting of palladium and nickel.
- 8. (AMENDED) A jewellery alloy according to claim 5[, claim 6 or claim 7], wherein the additional element is palladium and is present in an amount of between 0.5 wt% and 4.0 wt%.
- 9. (AMENDED) A jewellery alloy according to claim 5[, claim 6 or claim 7], wherein the additional element is nickel and is present in an amount of between 1.0 wt% and 2.0 wt%.
- 10. (AMENDED) An article comprising a metal component, wherein the metal component comprises a jewellery alloy according Claim 1 [to any one of the preceding claims].

- 13. (AMENDED) An [jewellery] alloy [containing] comprising 16.5 21.5 wt% aluminum, 0 4.0 wt% palladium, 0 2 wt% nickel and the balance gold [(except for impurities and incidental elements)].
- 14. (AMENDED) [An] <u>The</u> alloy <u>of claim 13</u> containing 18.5 19.5 wt% aluminum, 0.5 4.0 wt% palladium and <u>the</u> balance gold, <u>except for impurities and incidental elements</u>.
- 15. (AMENDED) [An] <u>The</u> alloy <u>of claim 13</u> containing 18.5 19.5 wt% aluminum, 1.0 2.0 wt% nickel and <u>the</u> balance gold, <u>except for impurities and incidental elements</u>.

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JEWELLERY ALLOY COMPOSITIONS

The present invention relates to novel jewellery alloy compositions.

Aluminium - gold alloys, with their comparable atomic size factors (2.878:2.8577), similar lattice crystal structure (f.c.c.) and large variation in electro-15 negativity factor, produce a diversity of microstructures and phases. The aluminium-gold phase diagram illustrates regions of solid solution, eutectic, and complex compounds (Au₅Al₃, Au₂Al, gamma, etc). The Au₂Al intermetallic compound is a complex cubic structure similar to ß 20 manganese and is a somewhat metastable state, with an electron: atom ratio of 3:2 and a weight percent ratio of 78.5%Au:21.5%Al. It is of particular interest to jewellers and the like because of its brilliant purple-golden colour. However, interest is largely offset by the fact that the 25 Au₃Al intermetallic compound is very brittle; like ordinary glass or porcelain it will fracture with a hard knock. In

fact, its brittleness is such that the Au, Al intermetallic

compound cannot be hardness tested using the Rockwell B

hardness testing machine with a 100 kg load; it will fracture even when a 60 kg load is applied.

According to the teachings of Japanese patent application JP 61-30642 in the name Tokuriki Honten Pte 5 Ltd, one way of overcoming the brittleness problem is to lower the gold component to 75 wt% whilst employing aluminium in an amount 20 to 24.5 wt%, and at the same time introducing 0.5 to 5 wt% of one or two additional elements selected from the group consisting of silicon, magnesium, 10 copper, zinc or manganese. By varying the relative amount of the additional element(s), the tone or hue of the colour may be changed subtly without losing the basic purple colour.

As can be seen from the Au-Al phase diagram, lowering the gold content below 78.5 wt% in the AuAl system gives rise to the co-existence of two structures - the Au₂Al intermetallic compound and the eutectic structure of Al and AuAl₃ - in the same sample. Thus, upon slow cooling from the molten phase or annealing of rapidly solidified samples, 20 precipitation of the aluminium rich eutectic phase on outward surfaces degrades the purple-golden colour. Even if rapidly solidified samples are not annealed, similar decolouration of the purple-gold colour may also occur after fabricating and polishing the jewellery and possibly 25 even through prolonged usage, albeit at a much slower rate. The hardness of the eutectic and Au₃Al phase is also significantly lower (around 10% for an alloy of 75 wt% gold and 25 wt% aluminium) than that of the Au₃Al intermetallic

compound. For these two reasons, the commercial viability of the alloy is limited.

It is an object of the present invention to provide a novel jewellery alloy which for the purposes of the present 5 specification is defined as having sufficient toughness to withstand Rockwell B hardness testing with a 100 kg load without shattering. Being able to use Rockwell B hardness testing is perceived as an empirical measure that the alloy is suitable for fabricating jewellery; if the alloy is too brittle to withstand Rockwell B hardness testing, it is too brittle to be used in jewellery. The term "jewellery" is intended to cover ornamental objects for personal adornment or otherwise, including medallions, and the like (eg coins) where the stated toughness is a prerequisite.

- In accordance with a first aspect of the present invention there is provided a jewellery alloy as hereinbefore defined, comprising 76-83.5 wt% gold and 16.5-21.5 wt% aluminium, and having a substantially purple hue (at least on annealing at 600°C).
- By definition, the jewellery alloy does not include pure intermetallic compound Au₃Al (78.5 wt% Au and 21.5 wt% Al) because it does not have the toughness to withstand Rockwell B hardness testing with a 100 kg load. The term 'substantially purple hue' includes the colours reddish or 25 pinkish purple and lighter purples.

Preferably, the hardness of the jewellery alloy remains substantially similar to that of the Au₃Al intermetallic compound; that is to say, the hardness of the

jewellery alloy is within about 6%, more preferably 5%, of the hardness of Au₃Al.

In one embodiment, the gold content may be above 78.5 wt% up to a maximum of 83.5 wt%, with the balance being aluminium. In this way, the requisite toughness is achieved by producing a gamma-phase gold aluminium structure.

In another embodiment, the jewellery alloy may have a gold content of less than 78.5 wt% and further comprise an 10 additional element selected from the group consisting of palladium and nickel. The aluminium content may preferably be 18.5-19.5wt%. The gold/aluminium ratio is preferably higher than 3.66. In preferred alloys, the amount of palladium when used as the additional element is in the 15 range 0.5wt% to 4.0wt%; the amount of nickel when used as the additional element is in the range 1.0wt% to 2.0wt%.

There is also provided an article comprising a metal component, wherein the metal component is fabricated from a 20 jewellery alloy in accordance with the present invention.

In accordance with a second aspect of the present invention, there is provided a jewellery alloy containing 16.5-21.5 wt% aluminium, 0-4.0 wt% palladium, 0-2 wt% 25 nickel and balance gold (except for impurities and incidental elements). The jewellery alloy may optionally contain small or trace amounts of elements, (eg oxygen) either constituting incidental constituents added in

accordance with established practice or present as impurities. In one embodiment, the jewellery alloy may be a binary alloy containing at least 16.5 wt% up to (but not including) 21.5% aluminium, and balance gold. In a second 5 embodiment, the jewellery alloy may contain 0.5-4.0 wt% palladium, with nickel substantially absent. In a third embodiment, the jewellery alloy may contain 1.0-2.0 wt% nickel, with palladium substantially absent. In all embodiments, the gold/aluminium ratio should be higher than 3.66. In the second and third embodiments, the aluminium content is preferably 18.5-19.5 wt%.

According to a third aspect of the present invention, there is provided an alloy containing 18.5-19.5 wt% aluminium, 0.5-4.0 wt% palladium and balance gold.

15 According to a fourth aspect of the present invention, there is provided an alloy containing 18.5-19.5 wt% aluminium, 1.0-2.0 wt% nickel and balance gold.

A better understanding of the present invention may be obtained in the light of the following examples embodying 20 the invention which are set forth to illustrate, but are not to be construed as limiting, the present invention.

Six example alloys embodying the present invention and two control alloys were manufactured and tested as follows:

1. All specimens were tested using a Rockwell B hardness testing machine with a 100 kg load. Where it was apparent that a specimen lacked sufficient toughness to withstand the Rockwell B hardness test, micro hardness testing with a 200g load was first

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conducted followed by an annealing and subsequent Rockwell B hardness testing.

ii) All specimens were annealed at 600°C and examined for precipitation of low melting point aluminium-rich eutectic. Such precipitation would be evident from the appearance of a greyish-white colour between reddish-purple regions on the specimen surface.

Control 1 (78.5 wt% Au and 21.5 wt% Al).

The Au₃Al intermetallic compound has a brilliant 10 purple hue, but is known to be brittle. The micro-hardness testing with a 200g load gave a reading of Vickers 250 (HRB-102 by conversion). After annealing no visible precipitates were found. Subsequent testing with Rockwell B hardness machine resulted in multiple fracturing of the 15 specimen.

Control 2 (75 wt% Au and 25 wt%Al).

The specimen has a reddish-purple colour, but was much softer than control 1 having a HRB of 91. Subsequent annealing resulted in large amounts of Al-rich eutectic 20 precipitation which seriously degrades the surface reddish-purple colour.

Example 1 (80.5 wt% Au and 19.5 wt% Al).

In comparison with control 1, the specimen was slightly softer (HRB of 101), but much tougher as 25 demonstrated by the fact that the sample survived Rockwell B hardness testing. Subsequent annealing showed no sign of precipitation and grain structure colour was pinkish-purple.

Example 2 (81 wt% Au and 19wt% Al).

In comparison with control 1, the specimen was softer (HRB of 96), but much tougher as demonstrated by withstanding a Rockwell B hardness test. Subsequent annealing showed no sign of precipitates and the grain structure colour was pinkish-purple.

Example 3 (79.7 wt% Au, 19.3 wt% Al and 1 wt% Pd).

In comparison with control 1, the specimen was 10 slightly harder (HRB of 103), but much tougher as demonstrated by withstanding a Rockwell B hardness test. Subsequent annealing showed no sign of precipitation and the grain structure was pinkish-purple.

Example 4 (79.7 wt% Au, 19.3 wt% Al and 1.0 wt% Ni)

- In comparison to control 1, the specimen was softer (HRB of 97.5), but much tougher as demonstrated by withstanding a Rockwell B hardness test. Subsequent annealing showed no sign of precipitates, and the grain structure colour was pinkish purple.
- 20 Example 5 (79.4 wt%, 18.6 wt% Al and 2.0 wt% Pd)

In comparison with control 1, the specimen was softer (HRB of 97), but much tougher as demonstrated by withstanding a Rockwell B hardness test. Subsequt annealing showed no sign of precipitation, and the grain 25 structure colour was pinkish purple.

Example 6 (77 wt% Au, 20 wt% Al and 3 wt% Pd).

In comparison with control 1, the specimen was slightly harder (HRB of 104.8), but much tougher as

demonstrated by withstanding the Rockwell B hardness test. Subsequent annealing showed no signs of precipitates and the grain structure colour was pinkish purple.

The foregoing examples demonstrate that it is possible 5 to make a tough purple gold-rich alloy by transforming the fragile and brittle Au₃Al intermetallic compound into the tougher gamma phase structure by either increasing the gold content above 78.5 wt% (75% molar content) or by alloying with additional element(s).

CLAIMS

- 1. A jewellery alloy as hereinbefore defined, comprising 76-83.5 wt% gold and 16.5-21.5 wt% aluminium, and having a substantially purple hue.
- 5 2. A jewellery alloy according to claim 1, having a hardness substantially similar to that of the intermetallic compound Au_3Al (78.5 wt% Au and 21.5 wt% Al).
- 3. A jewellery alloy according to claim 2, in which the hardness is within 6% of the hardness of the intermetallic 10 compound Au₃Al.
 - 4. A jewellery alloy according to any one of the preceding claims, consisting of more than 78.5wt% and up to and including 83.5 wt% gold and a balance of aluminium.
 - 5. A jewellery alloy according to any one of claims 1 to
- 15 3, further comprising an additional element selected from the group consisting of palladium and nickel.
 - 6. A jewellery alloy according to claim 5, in which the aluminium content is 18.5-19.5 wt%.
- 7. A jewellery alloy according to claim 6, wherein the 20 gold to aluminium ratio is at least 3.66.
 - 8. A jewellery alloy according to claim 5, claim 6 or claim 7, wherein the additional element is palladium and is present in an amount of between 0.5 wt% and 4.0 wt%.
 - 9. A jewellery alloy according to claim 5, claim 6 or
- 25 claim 7, wherein the additional element is nickel and is present in an amount of between 1.0 wt% and 2.0 wt%.
 - 10. An article comprising a metal component, wherein the metal component comprises a jewellery alloy according to

any one of the preceding claims.

- 11. An article according to claim 10, wherein the article is selected from the group consisting of ornamental jewellery, medallions and coins.
- 5 12. A jewellery alloy substantially as hereinbefore described with reference to the accompanying examples.
 - 13. A jewellery alloy containing 16.5-21.5 wt% aluminium, 0-4.0 wt% palladium, 0-2 wt% nickel and balance gold (except for impurities and incidental elements).
- 10 14. An alloy containing 18.5-19.5 wt% aluminium, 0.5-4.0 wt% palladium and balance gold.
 - 15. An alloy containing 18.5-19.5 wt% aluminium, 1.0-2.0 wt% nickel and balance gold

DECLARATION AND POWER OF ATTORNEY

#4

Atty. Dkt. No.: 5196-000003

DECLARATION

As a below named inventor, I hereby declare that:

My residence, mailing address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

JEWELLERY ALLOY COMPOSITIONS

the specification of which (check one)

| | is attached hereto. |
|-------------|---|
| | or |
| \boxtimes | was filed on July 31, 2001 as Application Serial No. 09/890,548 and |
| | was amended on (if applicable). |

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56, including for continuation-in-part applications, material information which became available between the filing date of the prior application and the national or PCT international filing date of the continuation-in-part application.

I hereby claim foreign priority benefits under 35 U.S.C. §§ 119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or any PCT international application having a filing date before that of the application on which priority is claimed:

| PRIOR FOREIGN APPLICATION(S) | | | | | |
|------------------------------|-----------|--------------|----------------|----|--|
| APPN. SERIAL NO. | COUNTRY | DATE FILED | PRIORITY CLAIM | | |
| | | (MM/DD/YYYY) | Yes | No | |
| 9900056-4 | Singapore | 02/02/1999 | \boxtimes | | |
| PCT/SG00/00013 | PCT | 01/31/2000 | \boxtimes | | |
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DECLARATION AND POWER OF ATTORNEY

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States provisional application(s) listed below:

| PRIOR PROVISIONAL APPLIÇATION(S) | | | |
|----------------------------------|-------------------------|--|--|
| APPN. SERIAL NO. | DATE FILED (MM/DD/YYYY) | | |
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I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s) listed below:

| PRIOR U.S. APPLICATION(S) | | | | |
|---------------------------|-------------------------|--|--|--|
| APPN. SERIAL NO. | DATE FILED (MM/DD/YYYY) | STATUS - PATENTED, PENDING, ABANDONED | | |
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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. § 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY



I hereby appoint Linda M. Deschere, Reg. No. <u>34,811</u>, of Harness, Dickey & Pierce, P.L.C., and each principal, attorney of counsel, associate and employee of Harness, Dickey & Pierce, P.L.C., who is a registered Patent Attorney, my attorney with full power of substitution and revocation, to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

CORRESPONDENCE ADDRESS

I request the Patent and Trademark Office to direct all correspondence and telephone calls relative to this application to <u>Harness</u>, <u>Dickey & Pierce</u>, P.L.C., <u>P. O. Box 828</u>, Bloomfield Hills, Michigan 48303 (248) 641-1600.

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DECLARATION AND POWER OF ATTORNEY

| Full name of sole or first inventor: Peng Chum LOH | |
|--|------|
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